The detection of trends in wet deposition data

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The Detection of Trends in Wet Deposition Data: Report of a Workshop

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Institute for Environmental Studies
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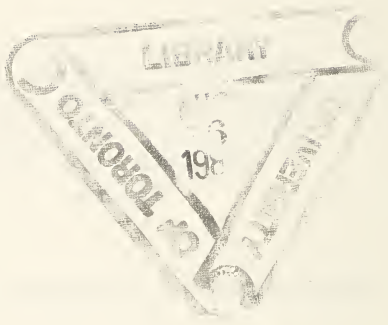

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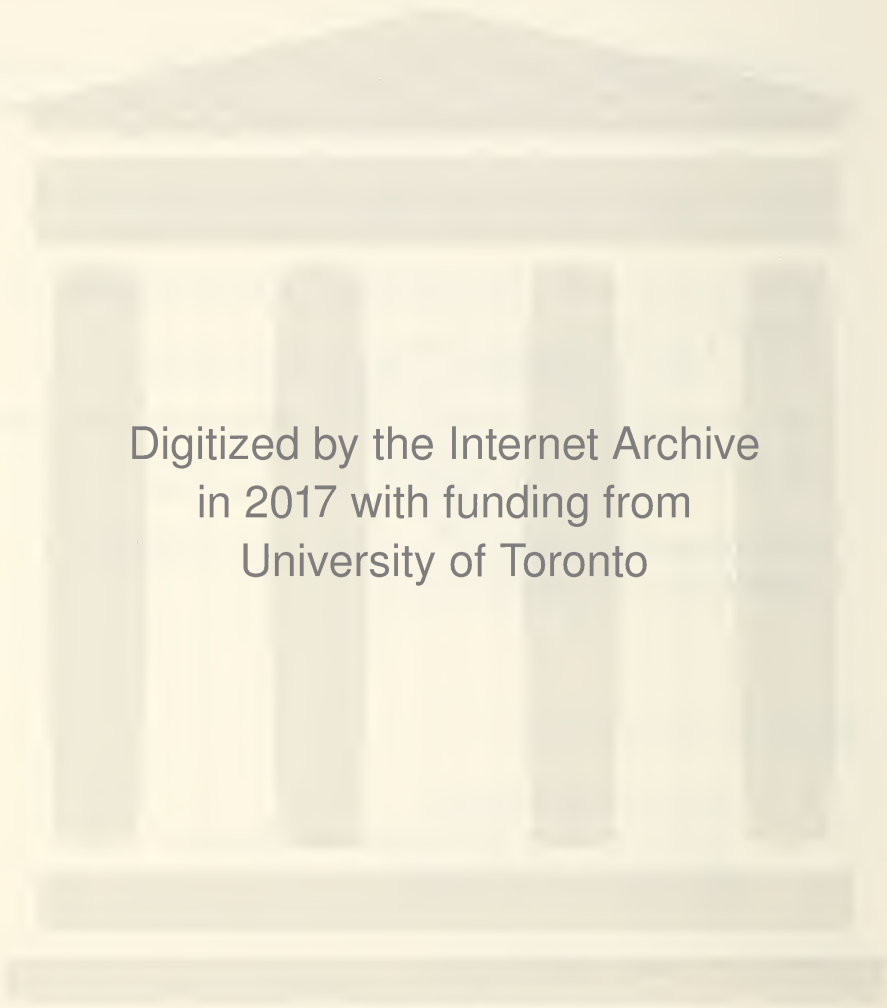

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Foreword

A Workshop on the Detection of Trends in Wet Deposition Data was held Nov. 17-18, 1983 at the Institute for Environmental Studies, University of Toronto. The emphasis was on methods rather than on results, and special attention was given to the question of estimating the length of time that would be required to detect a change in wet deposition resulting from some postulated change in regional emissions of air pollution. A background information paper had been distributed in advance, "The ability of wet deposition networks to detect changes in regional air pollution emissions" by R.E. Munn.

Based on material prepared at the workshop, a draft report of the meeting was prepared by the Rapporteur and circulated for comment. The suggestions received have been incorporated into this final draft.

Financial support by the Atmospheric Environment Service, Environment Canada is gratefully acknowledged. The Institute also wishes to thank the World Meteorological Organization and the United States National Oceanographic and Atmospheric Administration for providing travel funds for Dr. A. Eliassen (Oslo, Norway) and Mr. D. Pack (McLean, Va., U.S.A.), respectively.

The 20 Workshop participants are listed in Appendix 1.

1. Introduction[1].

A widely discussed question in the acidic deposition debate is the existence of trends and step changes in wet deposition data. This is the case not only with respect to historical time series, but also in relation to proposed control strategies: if emissions were to change by a designated amount, how long would it take to detect a corresponding change in wet deposition data?

Trend detection requires a statistical approach, with associated confidence intervals. However, because environmental time series are not well behaved, there is need for collaboration between statisticians and atmospheric scientists in the selection of appropriate methods. Some of the meteorological factors that should be considered in a statistical analysis are:

(1) Precipitation amount is a difficult meteorological measurement to make accurately, especially in gusty winds.

(2) Precipitation fields are characterized by large spatial gradients, particularly on the mesoscale in rough terrain and near coast-lines.

(3) Precipitation time series exhibit seasonal cycles (rain vs. snow; convective vs. frontal), against a backdrop of great random variability, with large spikes on some occasions but no rain on others. A rain-free period may last several weeks, for example.

(4) Mean values and frequency distributions of measurements of the chemical constituents of precipitation in different years sometimes do not appear to have been drawn from the same populations.

(5) Regional emissions of air pollutants are irregular in space and time. Even when the nearest strong point source is several hundred kilometres from a sampling station, wet deposition measurements could contain unusually large values on those rare occasions when plume dispersion was slight, the plume centre-line was directly overhead, and there was little precipitation between the source and the sampling site. (See Appendix 2.)

---

[1].The first draft of this section was written by P. Finkelstein and D. Whelpdale.

(6) The pathways of pollutants vary greatly from storm to storm and even from hour to hour within a single precipitation event.

In the general context given above, the focus of this report is directed toward methods of detecting future changes (lasting at least a year) in concentrations or deposition rates of the chemical constituents of precipitation. It would be particularly useful to have an idea in advance of the ease of detectability of changes in emission fields from time series of precipitation chemistry measurements. The space scale of most interest is about $10^4 km^2$ (deposition in an experimental watershed, for example) but changes over an entire region (e.g., eastern North America) are also important.

Specifically, the first two questions to be considered are statistical in nature:

1. In order to detect a change of a specified amount in wet deposition or concentration, what data are required (lengths of record before and after change; density of measurements)?

2. What is the minimum detectable change in wet deposition or concentration?

The next three questions pertain to the relationship between emissions and deposition in trend detection. Presumably changes in emissions are reflected in changes in the wet deposition and concentration fields. It is usual to use a model (conceptual, physical, mathematical; linear, non-linear) to relate the two. Hence, it may be possible to infer the impact of various emission control strategy scenarios on time series of wet deposition measurements.

3. How should emission changes be characterized or described statistically in order to optimize the chances of detecting resultant wet deposition and concentration changes?[2]

4. What is the spatial resolution of source regions that existing wet deposition networks are capable of distinguishing?

5. What are the implications of the above with respect to improving wet deposition networks?

---

[2].Changes in deposition/concentration should not be used to infer changes in emissions; more direct methods are available.

Finally there is a question relating to the fact that the main objective of an acidic deposition control strategy is to protect sensitive ecosystems; a detectable change in wet deposition or concentration is important only as one indicator in a biogeochemical chain stretching from sources to receptors.

6. Given a change in emissions, can a change be detected in sensitive soils, lakes and rivers and in the ecosystems they support?

Question 6 is beyond the scope of this report. With respect to the other five questions, the intent is to provide a framework for detailed studies rather than specific answers. The outstanding knowledge gaps will be discussed and some recommendations for priority studies that need to be undertaken will be made.

2. Reasonable scenarios for future changes in regional air pollution emissions[3]

2.1 Some general remarks

To provide a focus for discussion of the detectability of changes/trends, it is useful to begin by considering the kinds of pollution control strategies that might be adopted, and the resulting changes in regional emissions that might ensue. Of course it should be realized that countries will not all adopt the same control strategies and that emission reductions will not take place over-night following passage of legislation or administrative order. It should also be mentioned that the following remarks relate to sulphur in the context of Eastern North America; the situation with respect to other pollutants and other parts of the world may be quite different.

Actions have been underway for many years to control emissions of many pollutants, particularly oxides of sulfur and nitrogen. In the last decade, additional emphasis has been given to this question, particularly with respect to sulphur emissions from smelters and coal-fired power stations. Sulfur oxides contribute approximately two-thirds of the acidity in precipitation. Present and planned control actions, therefore, focus largely on the reduction of sulfur, primarily in the form of SO_2 gas.

---

[3].This section was written by D. Pack and R.W. Shaw.

2.2 Possible changes in emission patterns

The approach adopted here is to provide emission reduction scenarios that reflect reasonable expectations for the time required for the construction of emission control facilities and legal and administrative activities. These scenarios can then be used in a variety of simulations to examine the implications with respect to the detection of changes/trends in wet deposition.

(a) United States

Most contemplated SO_2 control actions in the United States focus on emissions from electrical generating stations burning fossil fuels containing sulfur. The sulfur oxide emissions resulting from electrical utilities in 1980 were estimated at 15.8 x 10^6 tonnes, representing 66% of the estimated total emissions of sulfur oxides in the United States in 1980 (MOI, 1982).

The exact nature or extent of sulfur oxide reductions in the U.S.A. has not yet been decided. Several proposals are being considered at the present time, including a mandatory coal-washing program, a ceiling on allowable SO_2 emissions of 2 lb/10^6 Btu (1 kg/10^9J), and the Sikorski-Waxman Bill (H.R. 4300) involving a 10 x 10^6 tonne reduction in SO_2 emissions (Streets et al., 1983). Generally speaking, the proposed emission reductions are not uniform, but vary geographically according to the nature of the sources, the availability of alternate fuels containing less sulfur, the applicability of abatement technology such as coal washing and scrubbers as well as economic and other factors. It is also possible that the atmospheric linkages between sources and receptors, as expressed by the source-receptor matrices generated by long-range atmospheric transport models, may be used to optimize spatially the emission reductions such that the minimum amount of sulfur is removed to achieve a given reduction in deposition (Young and Shaw, 1983). Such an optimized emission reduction would maximize deposition reductions which, in turn, would have the double benefit of maximizing the reduction in environmental stress and maximizing the ease of detecting it.

For the purpose of the analyses in this paper, we have assumed a spatially uniform percentage emission reduction that increases in time as follows:

A gradual emission reduction will begin within one to two years as readily-available techniques (e.g., coal washing, fuel switching, etc.) are brought on line by individual utilities. The reductions will then accelerate as very large sources complete the complex engineering systems required for SO_2 controls, then taper off until the total reduction is achieved in 10-15 years or so.

This scenario in the United States is shown in the left half of Table 2-1, both as a percentage of utility emissions and as a percentage of total United States emissions.

(b) Canada

In Canada in 1980, 45% of the 4.8×10^6 tonnes of sulfur dioxide emissions were from non-ferrous smelters and 16% from electrical utilities (MOI, 1982). Because the costs of controls at non-ferrous smelters are usually considerably smaller than those at thermal power stations ($100-1,000 per tonne sulfur at smelters compared to $1,000-4,000 per tonne at thermal power stations), it is likely that SO_2 controls in Canada would be implemented first at non-ferrous smelters, followed by measures at thermal power stations. A third possible measure, but not considered in this scenario because of the high cost involved, is reduction of emissions from residential and commercial fuel combustion by desulfurization of fuel oil prior to combustion.

Initial analysis using some of the long-range transport models considered by the Working Group under the United States-Canada Memorandum of Intent (Young and Shaw, 1983) indicates that the majority of emission reductions in Canada should take place in the provinces of Manitoba, Ontario and Quebec where the majority of non-ferrous smelters and thermal power stations are located. Meteorologically, it is most rational to abate emissions in these provinces because emitters there are located close to or upwind of sensitive receptor areas in Ontario, Quebec and the northern United States. It is estimated that abatement of sulfur emissions from smelters could take place in approximately 5-7 years, resulting in a 20% reduction in Canadian emissions, with control of utility emissions in southern Ontario in approximately 10 years, resulting in an additional 5% reduction in Canadian emissions. The chronology of emission reductions in Canada, and in North America as a whole, is also shown in Table 2-1.

TABLE 2-1

SULFUR OXIDE REDUCTION SCENARIOS (CUMULATIVE PERCENTAGE)

Time (Year After Action)	U.S.A. (Utilities Only)	U.S.A. (All Sources)	Canada (All Sources)	North America (% of all Emissions)
1	0	0	0	0
2	2	1	0	1
3	4	3	0	2
4	8	5	0	4
5	12	8	20	10
6	16	10	20	12
7	25	16	25	17
8	32	21	25	22
9	36	24	25	24
10	39	26	25	26
11	42	28	25	28

3. Sources of uncertainty in wet deposition and concentration data

The next main question that needs to be considered is the quality of wet deposition/concentration data sets, i.e., measurement precision, accuracy and representativeness.

3.1 Sampling and analytical errors at a single station[4]

Precision of wet deposition data can be determined from replicate sampling. (Here sampling includes handling.) Results are expressed in terms of coefficient of variation (COV = standard deviation/mean concentration) and they vary from network to network (See Table 3-1). Typical values for H^+, SO_4, NO_3^- and NH_4^+ from the Acidic Precipitation in Ontario (APIOS) network are of order 10% for daily sampling. Corresponding values for cumulative sampling over an averaging time of about a month may be somewhat higher. Table 3-1 also summarizes precision estimates from the Electric Power Research Institute (EPRI) precipitation network (Topel et al., 1982). Relative precision, instead of COV, is calculated. Relative precision is defined as the mean absolute difference between paired measurements divided by the median concentration of the observable. The calculated relative precision for H^+, SO_4 , NO_3^- and NH_4^+ for the EPRI network range from 5 to 9%. Precision is lower for soil-related species such as calcium. Seasonal dependence of sampling precision is expected to exist, although notreported.

Accuracy of wet deposition data is important. However, there is no real way to determine it. Employing standard solutions as a tracer for errors introduced in the chain of processes in sampling and analysis gives error estimates for what happens after the sample is collected. It does not address whether the right precipitation sample was collected nor does it address any potential chemical/physical changes that took place after collection.

Differences in data obtained from daily and cumulative sampling have been speculated to exist by some researchers. However, it should be borne in mind that the differences may be due to the lack of quality assurance in the earlier network data (monthly) and the improved quality assurance of the recent data

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[4].This subsection was written by W.H. Chan.

TABLE 3-1

Precision on precipitation concentration by wet-only sampling for some EPRI (Topel et al., 1982) and APIOS (Chan, 1984) comparisons. The APIOS daily results are based on triplicate sampling of 59 events; the APIOS 28-day results are based on triplicate sampling of 5 summer months. No information was given by Topel et al. (1982) on numbers of samples.

	Daily Sampling		
Parameter	EPRI	APIOS	28 Day Sampling APIOS
	(RP%)*	$(COV\%)_+$	$(COV\ \%)_+$
H^+	9	12	9
SO_4	5	6	10
NO_3^-	5	7	19
NH_4^+	8	10	9

*RP% = (mean absolute difference between pairs/median concentration) x 100%.

+COV% = (standard deviation/mean concentration) x 100%.

(daily) rather than due to differences in the sampling periods or protocols used. Madsen (1982) evaluated results from daily, weekly, and bi-weekly sampling intervals and concluded that the length of sampling interval under carefully controlled conditions appears to have little influence on determined concentrations of major anions and cations. It is also noted that data obtained from good quality-assured daily and monthly networks are comparable (Chan, 1984).

Analytical errors are in general small compared to sampling and handling errors (see for example, Topel et al., 1982).

It is important to implement good quality assurance programs in both field and laboratory practices.

3.2 Bias in network design[5]

In drawing conclusions for areas from measurements made at points, it is necessary to assume that the sampling sites (taken together) are representative of the larger area. In rugged terrain, however, monitoring stations are usually located at low altitudes, which obviously limits the degree to which they represent the entire study area. Similarly, important mesoscale precipitation and deposition gradients inland from coasts may be undetected by a macroscale network of stations. For this reason, model predictions (areal averages) may not agree with precipitation chemistry observations (point values).

The question of network bias due to sampling site location may be investigated by adding a few additional samplers in areas that are thought to be underrepresented in the existing network. For example, if one suspects that a particular network is biased because it contains only low-altitude stations, some ridge stations should be installed. Here the recommendation is made that statistical comparisons be undertaken of model-derived vs. areally-averaged actual wet deposition and concentration values for the same time period. (The actual values should be spatially smoothed (Thiessen polygons, for example) (Thiessen, 1911) before making the comparisons. This is required since even the most advanced models use area-averaged emissions and, usually, predict area-averaged deposition rates.)

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[5].The first draft of this subsection was written by M. Hawley, P. Summers and P. Cohen.

Finally, it will be necessary to determine the area that each sampling site represents. This could be done by variogram analysis (Matheron, 1963, 1967). Alternately, an estimate might be obtained by collecting meteorological data from several weather stations and using cluster or correlation analysis techniques to identify those stations where variations in the meteorological variables are similar to that observed at wet deposition sampling sites. In this case, however, it should be emphasized that there are a number of questions that must be investigated; for instance, which meteorological variables should be considered, and which clustering algorithm should be used?

3.3 Merging data from various networks[6]

There are numerous precipitation chemistry networks employing different sampling sites and protocols, and measuring different constituents of precipitation in some cases. Some of the networks in eastern North America are as follows:

Network	Number of Sites in Eastern North America	Length of Record (years)	Measurement Frequency
MAP3S	9	7	event
NADP	30 - >115	6	weekly
UAPSP	20	5	daily
APN	6	4	daily
CANSAP	25	6	monthly for old sets but now converting to daily

[6]. The first draft of this subsection was written by M. Hawley, P. Summers and P. Cohen.

Merging of data from two or more networks is an attractive idea because it increases the density of data points, permitting the construction of more accurate isopleths. This only is realized, however, if the networks are comparable. For an example of a successful merging of data from two networks, see Pack (1980).

In order to evaluate the feasibility of merging data sets, at least one common site should be chosen for operating the different types of network samplers. Six CANSAP and NADP samplers are co-located, for example.

Although various networks have different frequencies of measurement, it should be possible to aggregate event or daily data from one network to obtain weekly or monthly values that can be compared directly with data from other networks. In the case of pH, however, the numerical values depend in part on the length of time between the end of precipitation and the time of analysis. (The pH is affected by organic activity in the sample.) This may be true for other variables as well.

4. Variability and sources of uncertainty in the atmospheric processes[7]

Even if sampling, analytical and siting errors could be removed from wet deposition measurements, the data sets would still contain considerable variability due to meteorological influences. As an example, Fig. 4-1 shows the wet deposition of sulphate at Long Point, Ont. (on the north shore of Lake Erie) over a 14-month period. Most of the variability shown is caused by day-to-day changes in the weather patterns.

It might be thought that by increasing sample size to include several years of observations, mean values might approach some "true" values. However, meteorological variability extends over climatological time scales and wet deposition data from consecutive years may sometimes appear to have been drawn from different populations. Fig. 4-2 shows some results obtained by Samson et al. (1983) for years 1978, 1979 and 1980, based on a regional acidic deposition model and assuming an unchanging emission field. The figure compares the percent contribution to wet deposition of sulphur at Whiteface Mountain, New York from Ohio, Pennsylvania, New York and West Virginia, and shows significant interannual differences.

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[7].The first draft of this section was written by L. Barrie.

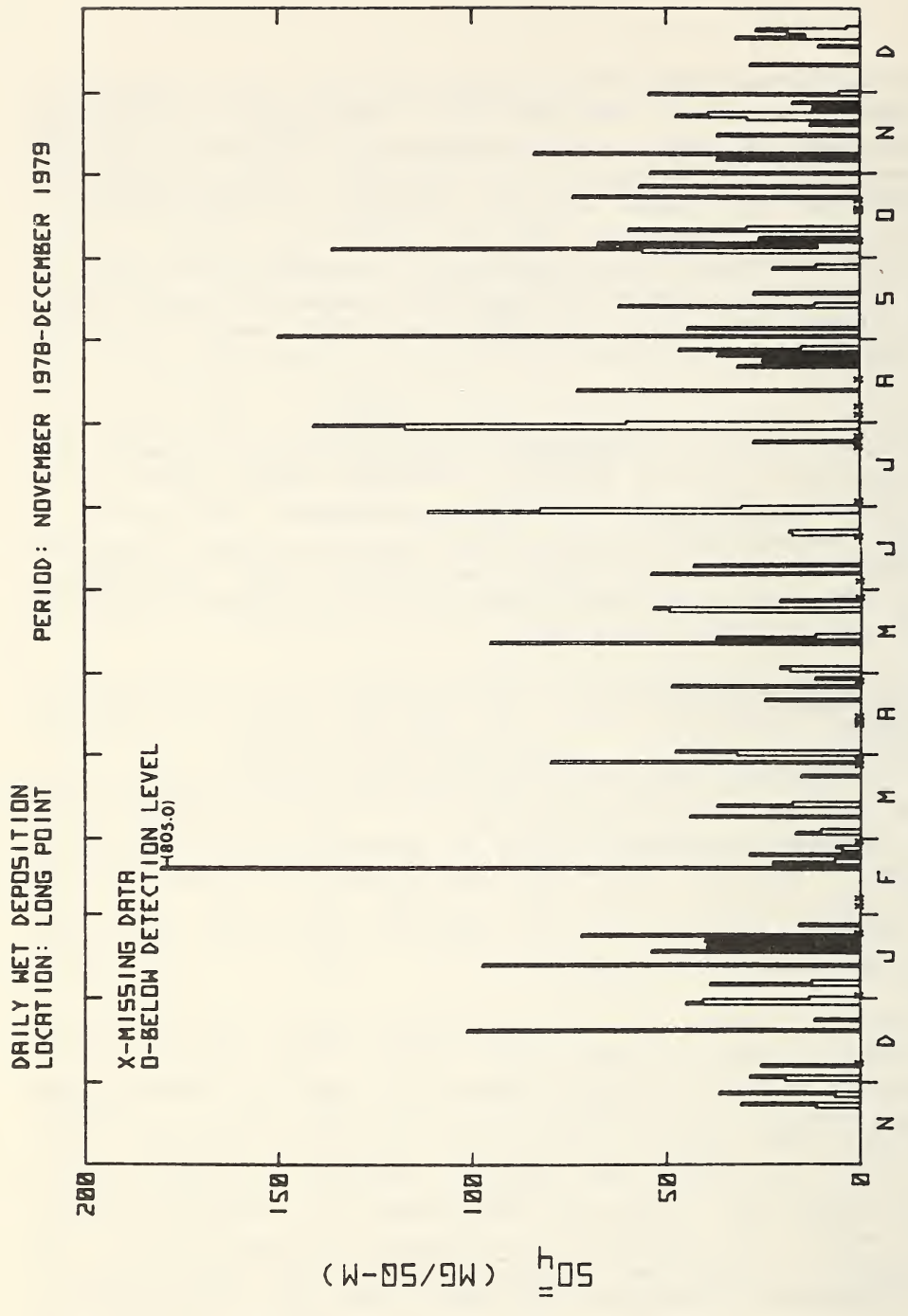

Fig. 4-1: Wet deposition of sulphate at Long Point, Ont. Nov. 1978 to Dec. 1979.

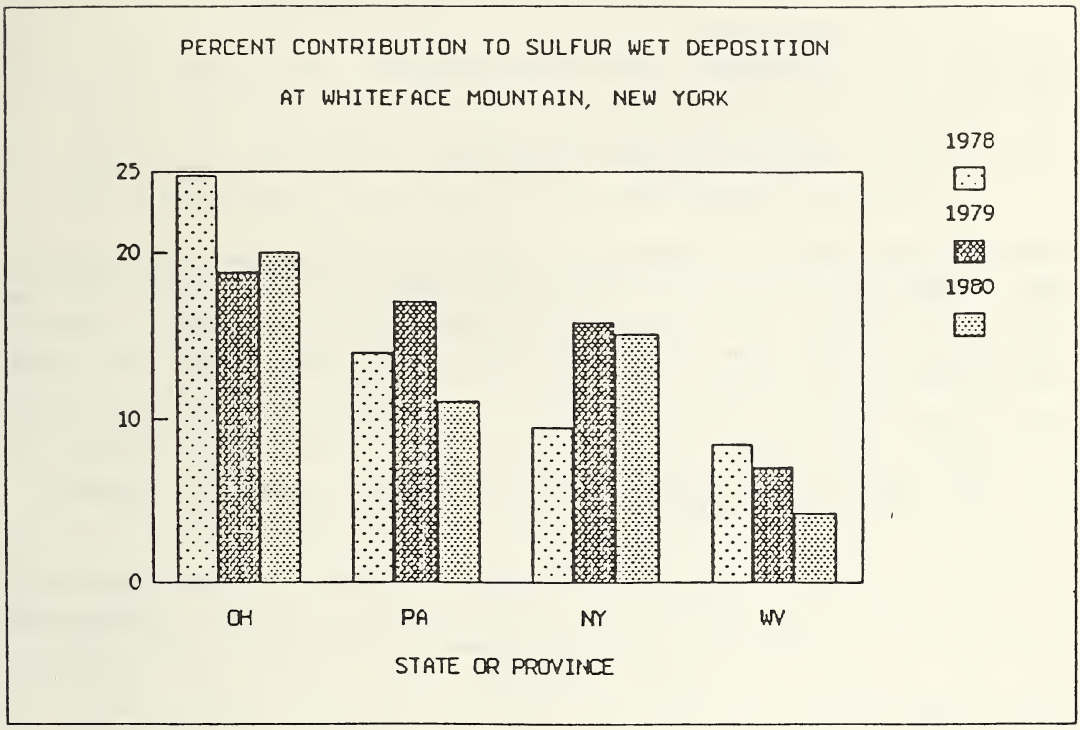

Figure 4-2: Calculated absolute contribution of four upwind States to sulfur wet deposition at Whiteface Mountain, New York over the period 1978 to 1980 (Samson et al., 1983).

The atmospheric factors that influence wet deposition and concentration levels are as follows:

1. Emissions
2. Transport and diffusion
3. Photochemistry
4. In-cloud chemistry
5. Precipitation scavenging
6. Dry deposition

Normalization of wet deposition time series insofar as possible with respect to these processes can improve our capability to detect the effect of emission reductions. One approach is sector analysis, in which the only cases considered are those with back-trajectories from designated sectors. Another technique, multiple discriminant and/or cluster analysis, can be used to subdivide climatological time seies into several groups. Then pre-intervention and post-intervention deposition data may be compared separately for each group. See Mueller and Hidy (1983), for example.

Simple normalization schemes are also possible, to remove the influence of a particularly wet or dry season or of a sequence of anomalous back-trajectories, for example. However, such efforts are not always successful because data sets are sometimes exceedingly noisy.

5. Methods for detecting change in wet deposition and concentration time series

5.1 Creation of "before" and "after" time series

In order to study the detectability of a change in wet deposition or concentration, two time series must be compared:

- one that is representative of conditions before some time T_o, the time when a change in regional emissions took place;
- the other that is representative of the new emission regime.

These time series can be obtained in two ways:

1. from historical data, selecting periods when regional emissions were higher or lower than usual:
   (a) during an industrial shut-down such as those that have occurred in Sudbury, Ontario. In this case, precise dates can be given to time T_o.

(b) by patching data from several years of high industrial activity with data from several years of regional economic slow-down.

2. from long-range transport models, changing the regional emissions at some time T_o using an emission control scenario like the one given in Section 2. In this connection, it should be mentioned that a model-derived "after" series should not be patched to a measured-derived "before" series.

In either case, it is desirable to test whether the selected periods are climatologically representative. Additionally, with respect to method 2 (modelling), it is desirable to test whether the choice of T_o makes any difference. (In the case of method 1, time T_o is fixed by the time of start-up of the industrial dispute.)

Before actually beginning the statistical analysis, the time series should be cleaned up, e.g., by combining event data to obtain monthly wet deposition or precipitation-weighted concentrations. Missing observations should be replaced by the ensemble mean, with outliers in the frequency distributions being examined separately. Other suggestions are given in Section 5.2 below.

5.2 Statistical Methods for Comparing "Before" and "After" Time Series[8]

In this section we discuss some of the statistical methodology which can be used in trend detection problems. It should be stated at the outset that this discussion is general, and not meant to be a recipe for all trend analysis problems. Each set of data must be treated individually, with changes in the statistical analysis as needed.

We begin by rephrasing the trend detectability question into more statistical language. Suppose that we want to test for trends at the 95% significance level, that we have X years of data, that the true (but unknown in practice) change in concentration (deposition) is Y, and that we want to have a Z% chance of declaring a statistically significant trend to be present. Detectability is usually assessed by fixing any two of X, Y, or Z, and determining the third. For example, suppose that we have 10 years of data with a known change, what is the probability of finding a statistically significant trend? Or, how many years of data are needed

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[8].This Section was written by G. Oehlert.

to have an 80% chance of finding a significant trend in data with a true trend of 3% per year? Of course, answers to these questions will vary from station to station, network to network, and constituent to constituent, with the principal determining factor being the size and statistical properties of the unexplained, and presumably random, behavior of the time series.

Before beginning the statistical analysis, the data must be prepared. This includes choosing which data to analyze, cleaning the data, choosing spatial and temporal scales, and making any necessary reexpression. We begin by considering only data from a single station; networkwide results are considered later. For the given station, we must choose which data to use. We can use all the data, or we can use only a subset, for example, only those data with back trajectories from an area with known emission changes. Such subsampling of the data can in theory increase the signal-to-noise ratio and make trend detection easier. We must choose a temporal scale. Some temporal aggregation is usually necessary to get regularly spaced data with few missing values, but excessive aggregation blurs the time series and can cause some loss of information. The proper time scale will vary from data set to data set. Finally, most statistical analyses make at least some assumptions about the distribution of the data, and a transformation of the data is often necessary to ensure that these assumptions are at least approximately satisfied. The logarithmic scale is often the scale of choice.

Having chosen and prepared our data, we now present a variety of methods for analyzing it.

5.2.1 A meteorological approach

The first method is to use a meteorological emission/deposition model which predicts correctly, at least on average. The predictions may be monthly, seasonal, or annual depending on the time resolution and accuracy of the model. The observed data at time t, D(t), are decomposed

$$D(t) = P(t) + e(t),$$

where P(t) is the model prediction, and e(t) is a mean zero stationary stochastic series of prediction errors.[9] Suppose that at some point an intervention occurs, such as regulations which reduce emissions. Using the new emissions, the model would presumably continue to predict with the same acuracy as before the intervention. However, to detect the change we work differently. After the intervention, we use current meteorology but continue to use the old, preintervention emissions in the model predictions, and redecompose the data as

$$D(t) = P(t) + w\ C(t) + e(t),$$

where C(t) is the approximate shape of the change in emissions (and the presumed shape of the change in the observed data) and w is a multiplier to be estimated from the data. The C(t) function is assumed to be zero prior to the intervention.

In this framework, D(t), P(t), C(t), and e(t) are known or computable up to the present time. We use statistical methods to determine the accuracy with which w can be estimated after an intervention occurs. This determination makes the assumption that the statistical properties of the e(t) series will not change after the intervention and that the shape of C(t) is known at least aproximately.

5.2.2. A statistical approach

A second approach to the problem is purely statistical. In this case, we do not use a meteorological model to produce a prediction series P(t), but instead produce a descriptive model based solely on the data. The observed data are decomposed

$$D(t) = B(t) + w\ C(t) + e(t),$$

where B(t) is a baseline description of the data. This description could be a constant, or a line, or something more complicated, for example, a line plus a seasonal effect. Usually there are some parameters in B(t) that must be estimated from the data. In this approach, like the first approach, D(t), B(t), C(t), and e(t) are

---

9. Meteorological models sometimes show bias so it will be necessary to check that e(t) is indeed a mean zero stationary stochastic time series.

known, hypothesized or estimable from the data, and the relevant information for determining how accurately w can be estimated from the data is taken from e(t) and the shape of C(t).

The choice between these two approaches is one of deciding for which stations and time scales the meteorological models predict better than the simple descriptive models. This will depend on the statistical properties of the error series e(t) and it does not seem possible to guess a priori which method will work better. Probably both should be tried, and the one with the smaller prediction error used.

With either model, we have

$$D(t) - (P(t) \text{ or } B(t)) = w\, C(t) + e(t),$$

where the left-hand side of the equation is observable and w must be estimated. If there are no outliers and the series of differences is fairly well behaved, then ordinary least-square regression will provide an adequate estimate of w. This estimate is not optimal, but its degree of nonoptimality is usually small. However, the usual standard error given for the ordinary least-square estimate of w can be seriously in error. This is because there is usually temporal association present in the error series e(t), and this temporal association increases the variability of the estimate of w.

There are several techniques available that will compensate for this underestimate of variability. Two will be mentioned here. (All methods make the implicit assumption that future errors will have the same statistical properties as past errors.) The first is to model the error series as an autoregressive process (AR), or perhaps an autoregressive moving average process (ARMA). See Box and Jenkins (1976). Once the series has been adequately modeled, it is possible to compute from the autocorrelation function of the process a more accurate estimate of the standard error of w. The second method is to Fourier transform the data, and estimate the spectrum of the error series (or equivalently, to lag weight the observed autocorrelations). The size of the low frequency terms relative to the mean spectrum determines a multiplier for the ordinary least-square variability estimate. See Bloomfield et al. (1983) for an example of the Fourier analysis, and Cunnold et al. (1983) for an example of the lag weight analysis.

In another statistical method, we sample from the distribution of past errors e(t) to simulate future errors. The synthetic series is then analyzed for trend. The simulation process is repeated a number of times, each time adding simulated errors to an assumed trend, and the fraction of time the trend is detected in the simulation is the estimate of future trend detectability.

5.2.3 Analysing changes in frequency distributions

The entire discussion so far has dealt with changes in the mean level of deposition (concentration), but other aspects of the data can also be examined. In some cases, the variability of the data could change after an intervention, and it might be easier to detect a change in the variability than a change in the mean. For example, a large point source might appear in the deposition records of local stations as scattered (depending on plume direction) episodes of acute deposition. A reduction in emissions at this point source might be seen more clearly in reduced variability or flattening of the peaks of deposition at nearby stations rather than as a change in the mean. We can also consider changes in the entire frequency distribution of depositions rather than just their average. In such cases, ridit analysis could be used to compare the frequency distributions from year to year. In this approach, we calculate frequency distribution of deposition for each year, and compare these by finding the chance that a random observation for one year is greater than a random observation from a reference distribution (usually the distribution for all years of data). These exceedance probabilities are then checked for trend, usually by regression. To do future extrapolation using ridit analysis, one could choose at random one of the previous year's distributions, subtract some constant from all the values, and use the difference as the extrapolated distribution. This is done several times, the reference distribution is formed, and ridits calculated. See, for example, Craig and Faulkenberry (1979).

Ridit analysis compares with mean analysis as follows. Ridits are only concerned with order, and not magnitude. If the data are such that outlying observations are fairly common, and thus would greatly inflate the standard errors in a mean analysis, then ridit analysis could be preferable since it doesn't weight the far tails as heavily as mean analysis. On the other hand, if the data are well behaved, mean analysis is probably more powerful.

The use of these alternate methods has some disadvantages. Basically, methods based on means not only seek to detect some change in the level of deposition or concentration, they also give some indication as to how large any change might be. The alternate

methods may be more sensitive in detecting a change, but they might not give an indication of the size of the change in useful units.

5.2.4 Network analysis

All of the methods discussed above have dealt with estimating a trend at a single station. However, we usually have a network of stations over a region, and we wish to get an overall estimate of how deposition (concentration) is changing. There are two ways of searching for networkwide trends. The first is to compute a trend separately at each of the stations in the network, and then combine the results from the different stations. The second is to combine the data from the stations by some sort of areal averaging, and then compute a trend from the areally averaged data. Both methods must take into account the fact that sites that are close in space, geography, meteorology, or other factors are likely to behave similarly, since they have some of the presumed causal factors in common.[10] Thus trends derived from neighbouring or related stations do not contain completely independent information, and this lack of independence must be accounted for.

The method of computing trends separately and then combining is probably preferable here. This is because stations will have been operating for different lengths of time, perhaps using different methods, and quite likely having e(t) series with differing statistical properties. In such cases, an easy and reasonable way to combine trends from different stations is to use a components-of-variance approach. In this approach, the trend estimate at each station is decomposed into several components representing various influences on the estimate. For example,

$$T(i,j) = N + R(j) + S(i,j),$$

where T(i,j) is the trend of the ith station in the jth region, N is a component common to all stations in the network, R(j) is a component common to all stations in the jth region, and S(i,j) is a station-specific component. This analysis produces an estimate of the network-wide term N and at least partially accounts for the spatial autocorrelation. See Reinsel et al. (1981).

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[10] Some recent unpublished studies by D.H. Pack suggest that there is a seasonal dependence of the degree of "closeness" of sites.

5.2.5 Some examples

We close this section with two examples demonstrating some of the statistical techniques proposed.

Example 1: A point-source problem

In this example, we have a large local point source more or less surrounded by a network of stations. Furthermore, there are no other major local sources, so that deposition other than that from the large local source is due to long-range transport and can be considered spatially homogeneous over the network. Thus the data at time t and station j, D(t,j), can be decomposed:

$$D(t,j) = L(t) + I(t,j)\ M(t),$$

where L(t) is the contribution from long-range transport at time t, M(t) is the effect of the local plume, and I(t,j) is an indicator function (0 or 1) indicating whether the plume was over station j at time t.

For this model, M can be as large as or larger than L, so that the series at each station can be quite variable, with high peaks, depending on the wind direction. This variability makes it difficult to detect a trend in M(t), even one as large as a total shutdown, when the station data are used separately. (See Appendix 2, for example.) In this situation, it may make sense to combine stations prior to doing trend estimation.

Consider the sum of all K station data series; call this sum D(t,+). Then

$$D(t,+) = K\ L(t) + M(t)$$

where we here assume that the local plume is over one and only one of the network stations. (This assumption is probably incorrect in practice, but it suffices for illustration in this example.) In the D(t,+) series, there is no variabilty due to plume direction, only the variability due to L and M. Depending on the relative sizes and variabilities of the L and M terms, the signal-to-noise ratio in the D(t,+) series can be several times as large as that in the individual station series, making trend detection considerably easier.

A second approach to this problem looks for a trend in the interstation differences. According to the model, all but one of the stations register L(t) and the other one registers L(t) + M(t). Thus the difference of the largest minus the smallest will be M(t). In practice, the L(t) term is not constant over the K stations in the network; there is some variability so that the difference is M(t) plus a (hopefully small) term representing sampling, analytical, and some spatial variability. However, this series of differences should show a marked change after a total shutdown.

Both of these approaches make use of the fact that the local signal is present in one and only one of the network records at a given time. There are doubtless many other ways to exploit this fact and turn the wind direction variability to advantage. The main point though is that it may be possible to circumvent a major source of variability by carefully choosing the data to be analyzed.

Example 2: Trend analysis at a station

In this example, we look at the question of trend detectability at a single station. This example is relevant for either the meteorological/emission prediction approach or the purely descriptive approach. The data have been decomposed as

$$D(t) - (B(t) \text{ or } P(t)) = w\ C(t) + e(t), \text{ or}$$

$$A(t) = w\ C(t) + e(t),$$

$$t = 1, 2, 3, \ldots, n.$$

A(t) and e(t) can be observed up till the present, and C(t) is hypothesized into the future.

In the case of residuals from a purely descriptive model, we will assume that any seasonal patterns present in the residuals have been removed. This can be done by subtracting from each January residual the average of all January residuals, etc., or analogous subtraction for seasons. For residuals from the meteorological model, there should be no seasonality in the residuals. If there is, it should be removed in the same way.

In the standard regression model, we have a vector A represented as the sum of a fixed vector C times an unknown constant w plus a vector of errors e. Assume that the errors have covariance matrix Σ. The ordinary least-square estimate of w is C'A/C'C, where an apostrophe indicates matrix transposition. This estimate

has variance $C'\Sigma C/C'C\hat{}2$, where $\hat{}$ indicates exponentiation. The variance of the estimate based on the assumption of uncorrelated errors is $\hat{\sigma}2/C'C$, where $\hat{\sigma}2$ is equal to the value on the diagonal of Σ. Thus the 'true' variance of the estimate is equal to the usual estimate of the variance based on the assumption of uncorrelated errors times an inflation factor $C'(\Sigma/\hat{\sigma}2)C/C'C$, or equivalently $C'RC/C'C$, where R is the autocorrelation matrix of the errors.

Note that R is the autocorrelation matrix for the entire time series. If we are to estimate trend detectability, we must make some assumptions about the future behaviour of R. We assume that R can be estimated from the data at hand, that is, from the e(t) up till the present.

How can R be estimated? R is a symmetric matrix that has at most n distinct values; each diagonal of R is constant. Thus we only need to estimate the n autocorrelations in the data. Assume for the moment that the e(t) series has mean zero. This will be true, for example, if A(t) has been deseasonalized. Then the natural estimates of the autocorrelations r(i), i = 0, 1, ... , n-1, are

$$r(i) = \sum_{j=i+1}^{n} e(j)e(j-i) / \sum_{J=1}^{n} e(j)e(j)$$

For technical reasons, these estimates of the autocorrelations cannot be used as is; they must be smoothed by using lag weights. The required degree of smoothing depends on n, the length of the time series.

The following smoother function seems reasonable. It is based on cosine weights and is not substantially different from most lag weight smoothers. All lag-weight smoothers depend on a parameter K, the maximum lag autocorrelation to be used in the analysis. It is difficult to specify K a priori, but a range of candidate K's is given as follows (Parzen, 1964). Take a smallest K to be an integer in the range 0.05n to 0.10n; take a medium K to be twice this smallest K and take a large K to be twice the medium K. Try the analysis with all three K's; hopefully, the results will be similar. If they are not, it is probably best to err on the side of conservatism and take the K which gives the biggest variance inflation factor, although in some situations it may be possible to determine that one of the smaller factors is more appropriate.

To calculate the lag weight estimates, first compute the r(i) values as above. Next, for each i, calculate f(i) according to the formula

$$f(i) = (1 + \cos(\pi i/K))/2 \ , \ i \leq K,$$

$$f(i) = 0, \qquad i > K.$$

Use the smoothed values r(i)f(i) in the autocorrelation matrix R to compute the variance inflation factor C'RC/C'C.

We will use two example sets of hypothetical sulfate data. These are shown in Figures 5.1 and 5.2. Both data sets should be considered to be residuals from either a descriptive or meteorological model; both are analyzed in the log scale; both have the same standard deviation of 0.36. (The coefficient of variation on the natural scale is approximately equal to the standard deviation on the log scale.) Neither data set shows much trend, but the second set has a tendency to have local drifts.

Since the data are on the log scale, we must also use a predicting series C(t) on the log scale. Figure 5.3 shows percent reductions similar to those given in Section 2 in log terms.

For the example given in Figure 5.1, n is 84 corresponding to 7 years of monthly data. We take K values of 7, 14, and 28. Lag weights for these K values are shown in Table 5.1 while Tables 5.2 and 5.3 give computed autocorrelations and smoothed autocorrelations for the first and second example sets, respectively. Tables 5.4 and 5.5 give the nominal standard deviation and the three adjusted standard deviations for the estimated coefficient of C(t) for several different lengths of data after an intervention. These two tables also give the probability of declaring that a statistically significant nonzero trend exists when the true coefficient of C(t) is one.

We see that the two series produce very different standard errors and probabilities of trend detection. This is due to the differing autocorrelations present. Most observed series will probably be more similar to the first than the second, but we must use procedures that will adjust the standard errors if autocorrelations are present.

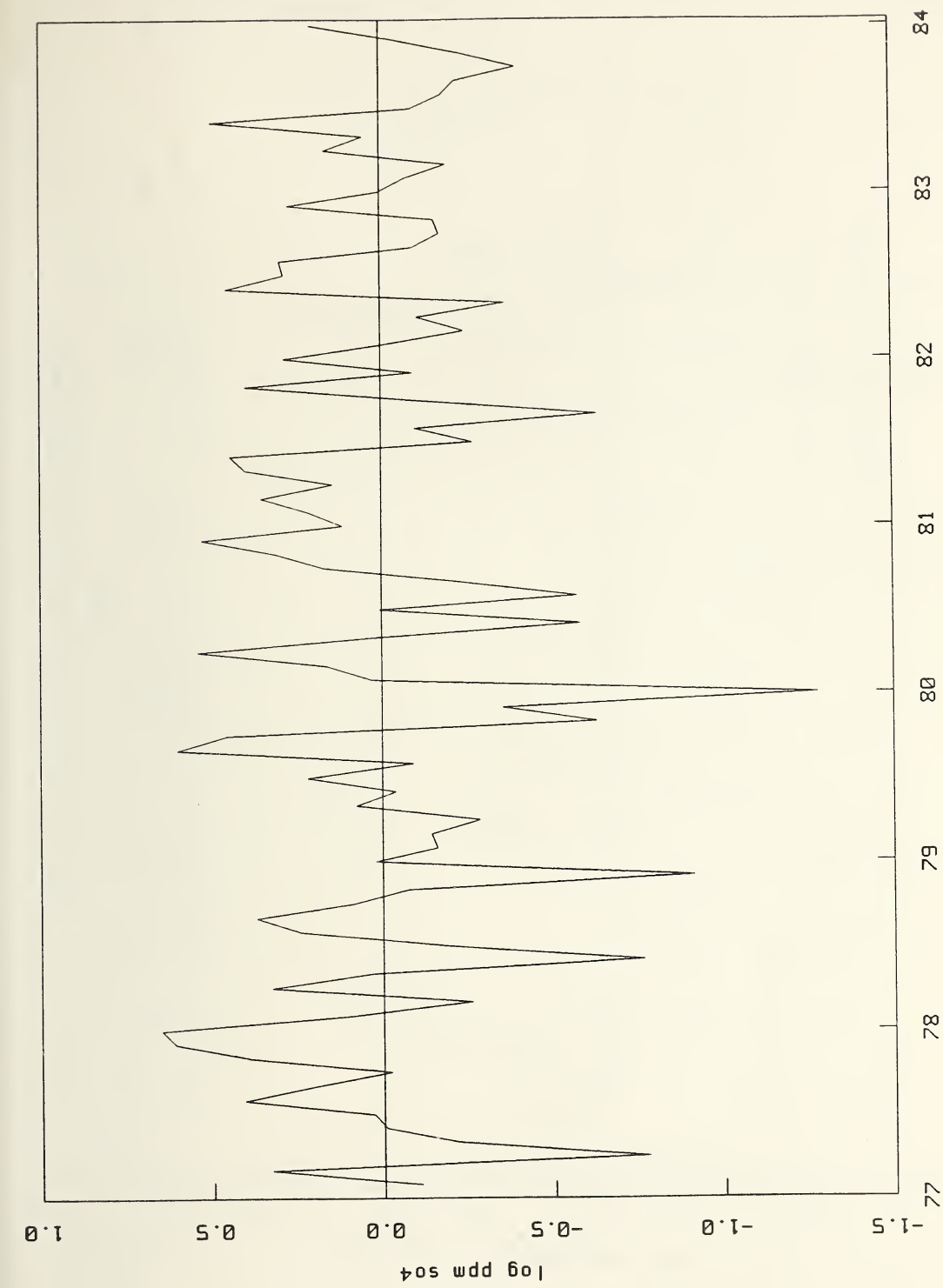

Fig. 5.1: Deseasonalized log sample SO_4 data, first sample.

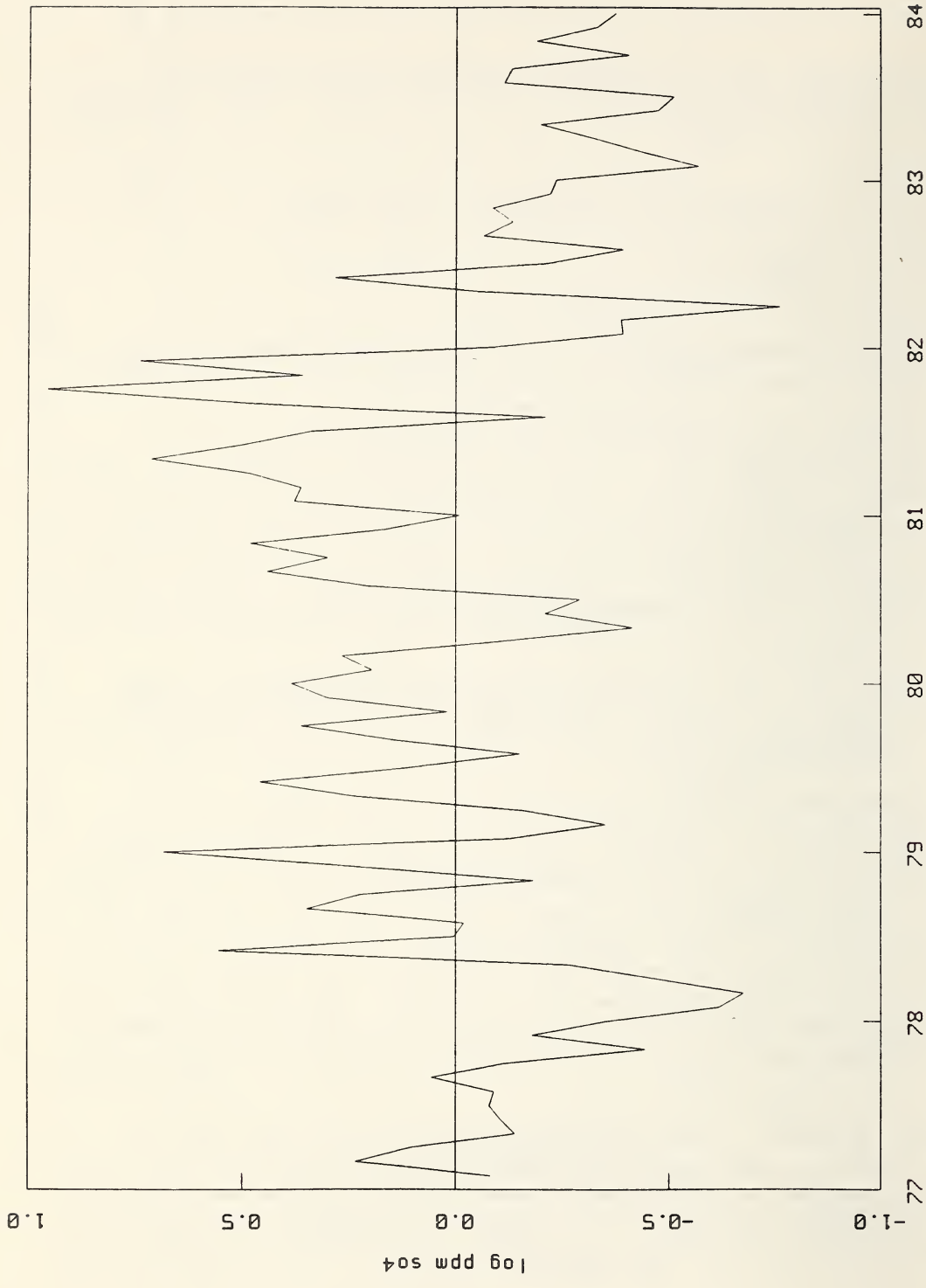

Fig. 5.2: Deseasonalized log SO_4 data, second sample.

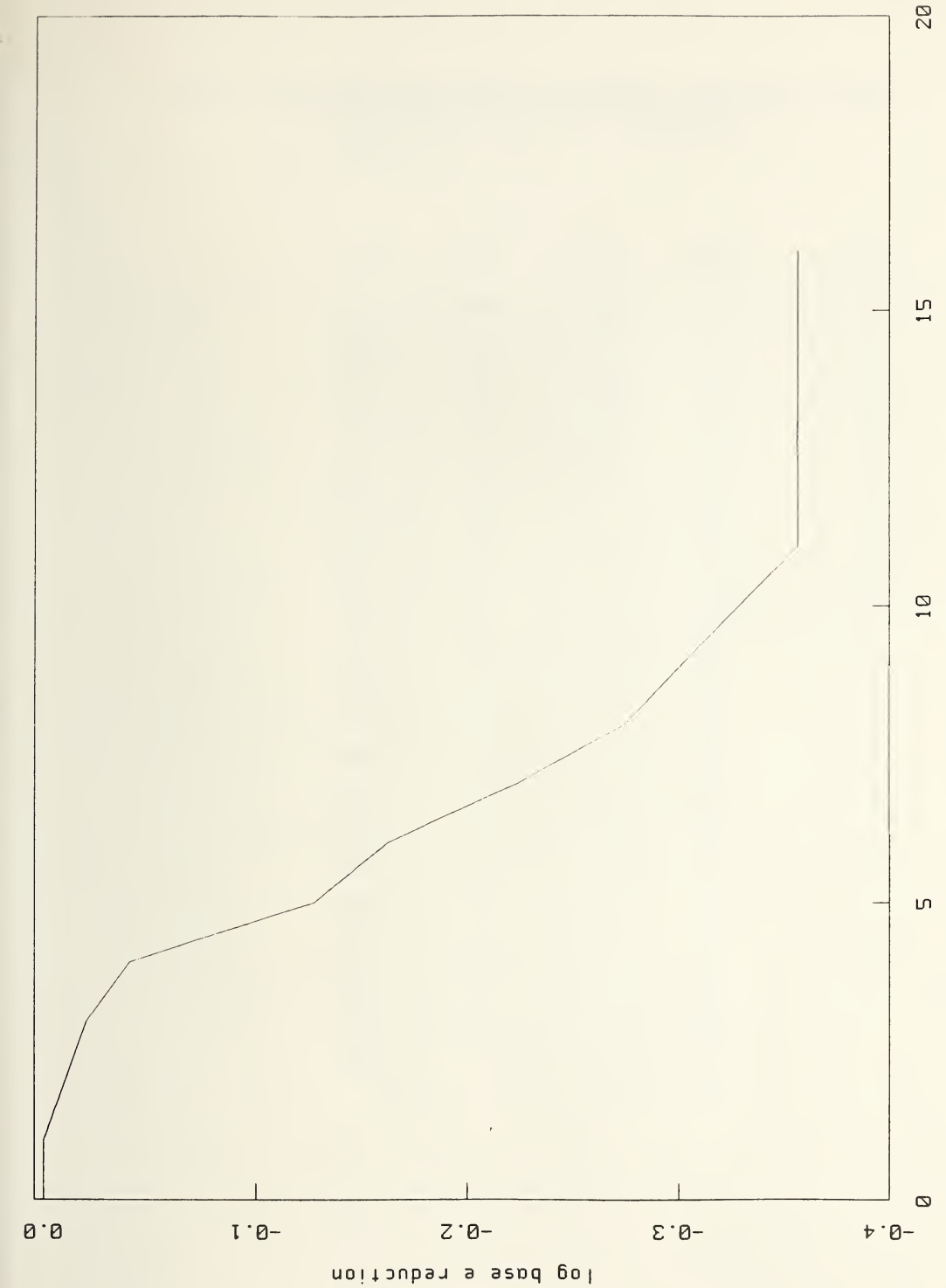

Fig. 5.3: Hypothesized reduction scenario.

Table 5.1: Lag weights for 28 lags and 3 K values.

	K		
lag	7	14	28
0	1.000	1.000	1.000
1	0.950	0.987	0.996
2	0.811	0.950	0.987
3	0.611	0.890	0.971
4	0.388	0.811	0.950
5	0.188	0.716	0.923
6	0.049	0.611	0.890
7	0.0	0.499	0.853
8	0.0	0.388	0.811
9	0.0	0.283	0.766
10	0.0	0.188	0.716
11	0.0	0.109	0.665
12	0.0	0.049	0.611
13	0.0	0.012	0.555
14	0.0	0.0	0.499
15	0.0	0.0	0.444
16	0.0	0.0	0.388
17	0.0	0.0	0.334
18	0.0	0.0	0.283
19	0.0	0.0	0.233
20	0.0	0.0	0.188
21	0.0	0.0	0.146
22	0.0	0.0	0.109
23	0.0	0.0	0.076
24	0.0	0.0	0.049
25	0.0	0.0	0.028
26	0.0	0.0	0.012
27	0.0	0.0	0.003
28	0.0	0.0	0.0

Table 5.2: Autocorrelations and lag weighted autocorrelations for the first sample data set.

0	1.000	1.000	1.000	1.000
1	0.225	0.214	0.222	0.224
2	0.020	0.016	0.019	0.020
3	-0.220	-0.134	-0.196	-0.213
4	-0.233	-0.091	-0.189	-0.221
5	0.027	0.005	0.020	0.025
6	-0.006	-0.001	-0.004	-0.006
7	0.051	0.0	0.025	0.043
8	0.047	0.0	0.018	0.038
9	-0.073	0.0	-0.021	-0.056
10	-0.103	0.0	-0.020	-0.074
11	-0.129	0.0	-0.015	-0.086
12	-0.060	0.0	-0.003	-0.037
13	0.165	0.0	0.002	0.091
14	0.043	0.0	0.0	0.021
15	-0.182	0.0	0.0	-0.081
16	-0.262	0.0	0.0	-0.102
17	-0.072	0.0	0.0	-0.025
18	0.180	0.0	0.0	0.051
19	0.192	0.0	0.0	0.045
20	0.289	0.0	0.0	0.054
21	0.009	0.0	0.0	0.001
22	-0.076	0.0	0.0	-0.009
23	-0.134	0.0	0.0	-0.011
24	-0.224	0.0	0.0	-0.012
25	-0.056	0.0	0.0	-0.002
26	0.049	0.0	0.0	0.0
27	0.070	0.0	0.0	0.0
28	0.035	0.0	0.0	0.0

Table 5.3: Autocorrelations and lag weighted autocorrelations for the second sample data set.

0	1.000	1.000	1.000	1.000
1	0.581	0.552	0.574	0.579
2	0.299	0.242	0.284	0.295
3	0.288	0.176	0.256	0.280
4	0.182	0.070	0.148	0.173
5	0.226	0.042	0.162	0.209
6	0.245	0.012	0.150	0.218
7	0.209	0.0	0.104	0.179
8	0.199	0.0	0.077	0.162
9	0.071	0.0	0.020	0.054
10	-0.032	0.0	-0.006	-0.023
11	0.020	0.0	0.002	0.013
12	0.087	0.0	0.004	0.053
13	0.088	0.0	0.001	0.049
14	0.028	0.0	0.0	0.014
15	0.040	0.0	0.0	0.018
16	-0.066	0.0	0.0	-0.026
17	-0.168	0.0	0.0	-0.057
18	-0.137	0.0	0.0	-0.039
19	-0.155	0.0	0.0	-0.037
20	-0.082	0.0	0.0	-0.016
21	0.008	0.0	0.0	0.001
22	-0.069	0.0	0.0	-0.008
23	-0.124	0.0	0.0	-0.010
24	-0.165	0.0	0.0	-0.009
25	-0.203	0.0	0.0	-0.006
26	-0.113	0.0	0.0	-0.002
27	-0.079	0.0	0.0	-0.001
28	-0.058	0.0	0.0	0.0

Table 5.4

Nominal and adjusted standard deviations

First data set

Years of data	nominal sd	K 7	K 14	K 28	probability of detection
4	2.87	2.97	2.65	2.32	0.05
6	0.59	0.61	0.53	0.45	0.36
8	0.29	0.29	0.25	0.20	0.92
10	0.18	0.19	0.16	0.13	0.99
12	0.14	0.14	0.12	0.09	0.99

Table 5.5

Nominal and adjusted standard deviations

Second data set

Years of data	nominal sd	K 7	K 14	K 28	probability of detection
4	2.87	4.94	51.71	6.06	0.03
6	0.59	1.02	1.19	1.27	0.11
8	0.29	0.50	0.59	0.63	0.34
10	0.18	0.33	0.39	0.41	0.67
12	0.14	0.25	0.29	0.31	0.89

6. Network optimization for detecting trends[11]

Let us assume that the observed deposition can be written as,

$$D_o = \langle D_o \rangle + e \qquad (7)$$

The ensemble averaging operator $\langle\rangle$ is defined with respect to a fixed emission field. The residual e between D_o and $\langle D_o \rangle$ is a function of the meteorology and chemistry that govern D_o. The value of $\langle D_o \rangle$ changes in response to emission variation. Trend analysis consists of detecting changes in $\langle D_o \rangle$ in the midst of the "noise" e. The general approach is to postulate crude models for $\langle D_o \rangle$ and e and fit them to data. We know enough about long-range transport processes to formulate models for $\langle D_o \rangle$ that represent big improvements over currently used statistical models.

It is obvious that trends are best detected where the signal $\langle D_o \rangle$ (related to emission change)-to-noise (e^2) ratio is highest. Wigley and Jones (1981) have used this concept in detecting temperature trends associated with increases in CO_2 levels. In order to estimate e^2, we have to model $\langle D_o \rangle$. However, if e^2 is much larger than the variance of $\langle D_o \rangle$ (associated with emission changes), one can calculate e^2 from observations of deposition assuming that $\langle D_o \rangle$ is constant. Then $\langle D_o \rangle$ is a simple average over the time series, if the series is sufficiently long. This procedure allows one to plot isopleths of signal-to-noise ratios for different seasons. The first indication of trends in the time series should be given by data from monitoring sites placed in the 'highs' of these isopleths. In principle, the signal-to-noise technique should be useful in selecting from amongst existing monitoring stations and in siting new stations. In practice, the field of signal-to-noise ratios may be rather chaotic, and it is recommended that appropriate statistical studies be undertaken to estimate the length of time required to obtain stable results.

Other methods should of course be applied in the search for optimal networks for trend detection, and the results compared. One approach that should particularly be mentioned is Kriging, which utilizes the spatial correlations that exist in a wet deposition field. See, for example, Finkelstein and Seilkop (1981).

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[11]. This section was written by A. Venkatram.

7. Alternative indicators of changes in regional emissions[12]

Up to this point, the assumption has been made that trend detectability is to be examined using wet deposition or concentration time series. There remain two additional questions to be considered:

1. Is wet deposition amount or is concentration in precipitation a more useful indicator of changes in regional emissions?

2. At some wet deposition observing stations, daily concentration of SO_2 and of sulphates in total suspended particulate matter are also monitored. Would these time series provide a better indicator of change than would the wet deposition data?

7.1 Precipitation concentration data versus deposition data

In order to answer the first question, data from four stations in the APN Network operated by the Canadian Atmospheric Environment Service have been examined for the years 1980 and 1981. The four sites are Kenora, Chalk River and Long Point in Ontario and Kejimkujic National Park in Nova Scotia. From these data, published by Barrie et al. (1982), Table 7.1 was constructed. The left-hand side of the Table covers daily precipitation observations, the right-hand side gives air concentrations (to be discussed in Section 7.2).

The Table shows that sufficient precipitation for an accurate chemical analysis was collected on about 100 days per year. The number of days with precipitation falling was in fact slightly larger than this, i.e., about one day in three. Except for one year (ELA-Kenora in 1981), the annual coefficient of variance (COV) for precipitation is very stable at between 100 and 120%. The next four columns show the annual arithmetic mean value and the COV for the measured $SO_4^=$ concentration in the rain and the computed SO_4 deposition (sum of the products of rain amount times $SO_4^=$ concentration for each event). This shows two interesting features - first, that in general the COV for the $SO_4^=$ concentration is less than that for the $SO_4^=$ deposition and thus, since n is the same for each of the concentration times series, should give an earlier indication of change. Second, there appears to be an inverse correlation between both the concentration and the deposition annual means and the COV as illustrated in Fig. 7-1a.

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[12].This section was written by P. Summers.

SITE	YEAR	PRECIPITATION DATA						AIR CONCENTRATIONS					
				$SO_4^=$ conc		$SO_4^=$ dep		SO_2			$SO_4^=$		
		No. of days with conc. data (n)	Precip COV (%)	Mean ($mg\ \ell^{-1}$)	COV (%)	Mean ($g\ m^{-2}$)	COV (%)	No. of obs. (n)	Mean ($\mu g\ m^{-3}$)	COV (%)	No. of obs. (n)	Mean ($\mu g\ m^{-3}$)	COV (%)
CHALK RIVER	1980	142	114	3.82	85	1.88	114	294	9.17	109	308	2.38	102
	1981	122	120	4.61	94	2.33	121	311	9.79	125	241	2.23	98
ELA-KENORA	1980	91	117	1.47	87	0.47	103	190	2.35	85	271	1.30	79
	1981	101	174	2.02	109	0.67	136	146	2.31	71	306	1.50	102
KEJIMKUJIC	1980	73	100	2.62	95	2.06	106	208	2.72	90	308	2.88	108
	1981	126	116	2.10	101	1.67	117	225	2.65	81	290	2.20	81
LONG POINT	1980	99	110	4.67	65	3.46	109	323	17.15	88	339	5.22	89
	1981	79	114	5.53	54	4.88	94	352	16.68	117	349	5.08	109

Table 7-1: Yearly values of the Coefficient of Variation COV (Standard Deviation divided by the Arithmentic Annual Mean) for each of four Canadian APN Monitoring Sites.

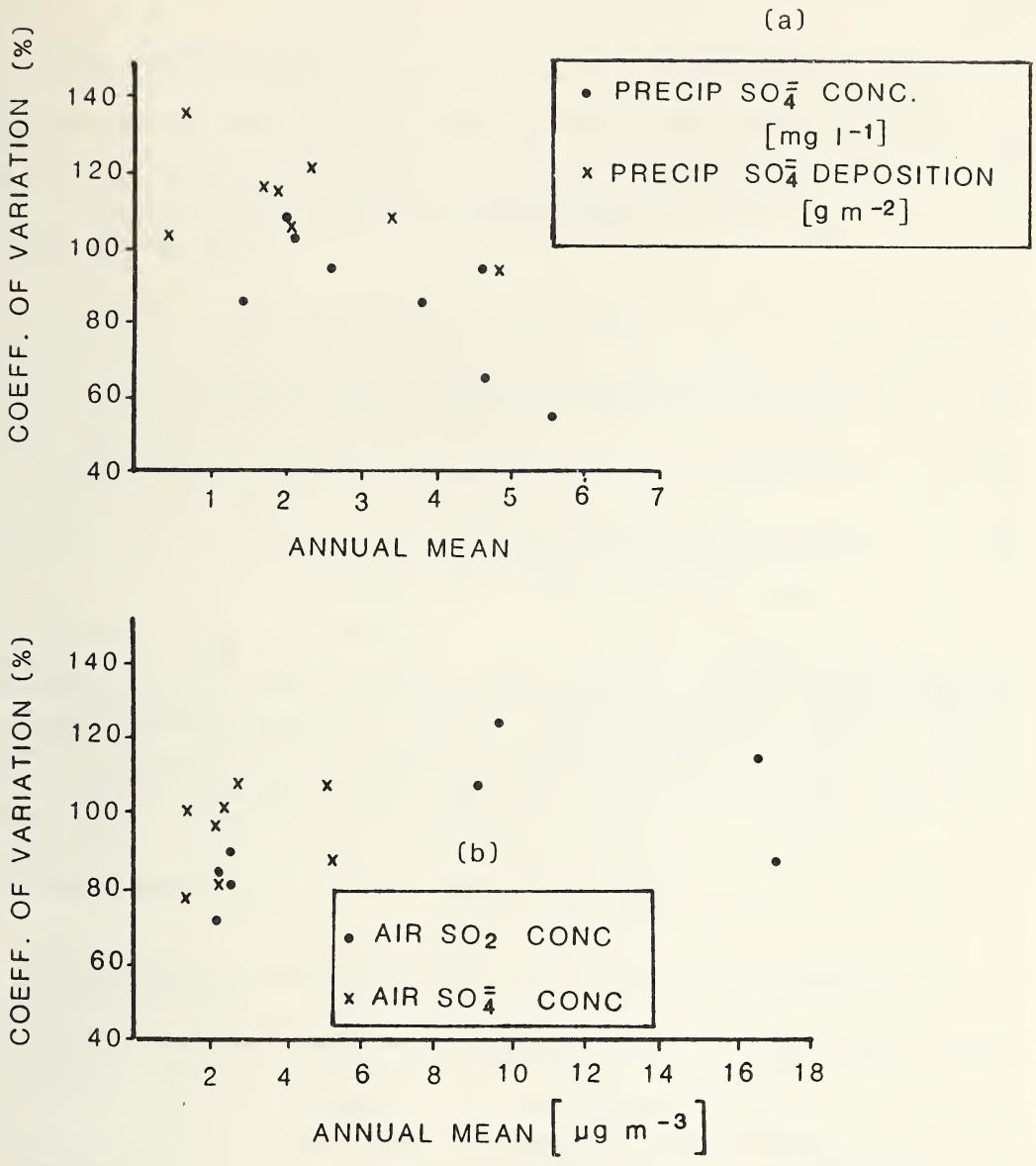

Fig. 7-1: Relationship between the coefficient of variance and the annual mean value for four parameters.

7.2 Air concentrations

The wet deposition (D) at a sampling site can be expressed as

$$D = f[SO_2, SO_4^{=}, R, X, X_2, \ldots]$$

where

SO_2 = ambient air concentration of SO_2 at the site

$SO_4^{=}$ = ambient air concentration of $SO_4^{=}$ at the site

R = precipitation amount/rate

X_1, X_2 = other meteorological variables.

Whereas values of D can only be obtained for days on which precipitation occurs - about one day in three in eastern North America - values of SO_2 and $SO_4^{=}$ are available every day (except when they are below the detection limits of the monitoring technique). Thus daily air concentration measurements could generate a time series of a given size in one third of the time and, if the coefficient of variation were approximately the same as for the deposition series, a trend could be detected much sooner.

The second question can now be investigated by considering the air concentration data from the four sites as shown in the right hand side of Table 7-1. The Table shows that for SO_2 and $SO_4^{=}$ concentrations, there are almost three times as many data points in one year as for $SO_4^{=}$ deposition. In general, the COV for SO_2 and $SO_4^{=}$ air concentrations are in the same range and both span the range of COV for the precipitation data. Thus, the advantage of using air concentrations data lies in the fact that, over a given period of years, the time series will contain about three times as many data points (n) as the equivalent series for precipitation data. This means that, in a given time period, the chances of detecting a statistically significant change in air concentrations are greater than for precipitation data. Or alternatively, because data points are accumulated faster, a change is likely to be detected sooner in the air concentration series.

From Fig. 7-1b, there is not such a strong relationship between the COV and the annual mean value, but if there is any trend at all, it is the opposite of that in Fig. 7-1a. In other words, for air concentrations, there is a tendency for the COV to increase as the mean concentrations increase.

7.3 Some general remarks

The above preliminary analysis shows that at all sites, the $SO_4^{=}$ precipitation concentration data are less variable than the $SO_4^{=}$ precipitation deposition data and hence, the former could be used as an earlier indicator of a statistically significant change due to emission changes.

Looking at the relationships in Figs. 7-1a and 7-1b together suggests that there is no single "best" parameter for detecting changes or trends. Precipitation concentration appears to be better than precipitation deposition especially where the mean values are high (i.e., nearest to the sources of SO_2). However, air concentrations have the advantage of the larger n and this advantage is maximized at low mean concentrations (i.e., farthest from the source of SO_2). The overall advantage of using air data at all locations needs to be investigated further.

Clearly then, an analysis aimed at detecting a change in wet deposition of $SO_4^{=}$ should examine other related observations such as $SO_4^{=}$ concentrations in precipitation and air and SO_2 concentrations in air where they are available. In this way, an interim indication of a trend due to emission changes may be detected whilst awaiting the possibly longer time required to confirm a change in the wet deposition.

8. Recommendations

A. With respect to measurements of the chemical constituents of precipitation

A.1 It is important to try to establish the precision and accuracy of wet chemistry data.

A.2 It is important to maintain existing precipitation chemistry networks, and to undertake periodic network intercomparisons of sampling and quality assurance protocols.

A.3 It is recommended that appropriate governmental bodies produce values (daily, monthly and annual means) of wet deposition and concentration areally averaged over sensitive areas such as Muskoka. These data are needed for comparisons with model-derived values.

A.4 In sensitive areas subject to mesoscale precipitation effects, experimental studies should be carried out, the objective being to improve current methods of estimating area-wide averages.

A.5 Existing networks should be examined with respect to their ability to detect the effects of a change in regional emissions. The examination could be accomplished through signal-to-noise or Kriging techniques, for example. Because regional emissions are not likely to change much during the next 5 years according to the sulphur emission scenarios given in Section 2, there is still time to expand the capability of existing networks for trend detection.) This recommendation should be given high priority.

B. General recommendation with respect to trend detection

Various statistical and modelling techniques are available for estimating the length of record that would be required to detect step changes and trends in wet deposition/concentration time series. A recommended strategy is to select several of the more promising techniques, bearing in mind the context of the investigation and the data sets available, and to compare the results. Qualitative agreement would lend credibility to the findings. As suggested in Section 7, a change in regional emissions will be easier to detect from a time series of precipitation-weighted concentrations than from the corresponding series of wet deposition values.

C With respect to long-range transport modelling

C.1 Better emission inventories are required, with at least week-to-week resolution in the case of the utilities.

C.2 For specific sensitive receptor areas such as Muskoka, it would be useful to try to estimate the fluxes into and out of the areas as well as wet and dry deposition. (Although this would be of great research interest, the estimates might not be of sufficiently high quality to be useful in an operational way.)

C.3 Better information is required on year-to-year variability in climate insofar as it influences wet deposition. How should a climatologically representative period be defined?

C.4 With respect to "before" and "after" time series joined at time T_o, empirical tests should be carried out to determine the range of results to be expected for differing choices of T_o.

C.5 Statistical comparisons should be undertaken of the properties of model-derived vs. observed wet deposition/concentration time series. (The observed time series should be averaged over grid squares with the same spatial resolution as the model outputs.)

D With respect to other indicators of a change in emissions

D.1 Daily measurements are available of SO_2 in air and SO_4 in total suspended particulate matter at some rural stations. As suggested in Section 7, these time series may provide better indicators of change/trend than do the wet deposition measurements.

D.2 Of the several chemical constituents of precipitation, some will be affected by proposed emission control strategies but others will not. It is therefore recommended that trend detection studies be conducted on a suite of substances, and the results compared.

9. Conclusion

This report provides advice to persons interested in estimating the length of a wet deposition time series that would be required to detect a change/trend in regional emissions. The six questions posed in the introduction (see pgs. 2-3) have not been answered quantitatively: to do so would require an analysis of observational data and/or of model results. However, the formulation of explicit questions was useful in focusing the discussion, which produced a number of suggestions on approaches that should be tried.

A set of recommendations has been given in Section 8, and there is need here for only a few additional remarks.

1. A percentage emission reduction that is constant over a large geographical area will be easier to detect than a spatially variable reduction.

2. Substantial gains in the ability to detect a change in regional emissions may be obtained by including information derived from meteorological models.

3. Use of a time series at a single station ignores a substantial amount of information. It may be possible to increase the probability of detection by examining information from several stations. One approach is to complete a trend analysis for each site individually and then combine the results, accounting for spatial correlation, for an overall test for trend. An alternative approach is to average spatially before considering the problem of trend detection. Using annual average data, a spatial estimation technique, e.g. Kriging, can be used to obain annual areal averages. Statistical approaches used for a single receptor can then be applied.

References

Barrie, L.A., H.A. Wiebe, K. Anlauf and P. Fellin (1982). "Results of the Canadian Air and Precipitation Monitoring Network APN: July 1980-Dec. 1981. Report AQRB-82-009-T, Atmos. Envir. Serv., Toronto.

Bloomfield, P., Oehlert, G., Thompson, M.L., and Zeger, S. (1983). A frequency domain analysis of trends in Dobson total ozone records, J. Geophys. Res. 88, 8512-8522.

Box, G.E.P. and Jenkins, G.M. (1976). Time Series Analysis, Forecasting and Control, Holden-Day, San Francisco.

Chan, W.H. (1984). Unpublished results, Ont. Min. of the Envir.

Craig, C.D. and Faulkenberry, G.I. (1979). The application of ridit analysis to detect trends in visibility. Atm. Env. 13, 1617-1622.

Cunnold, D.G., R.G. Prinn, R.A. Rasmussen, P.G. Simmonds, F.N. Alyea, C.A. Cardeline, A.J. Crawford, P.J. Fraser and R.D. Rosen (1983). The Atmospheric Lifetime Experiment 3. Lifetime methodology and application to three years of CFC13 data, J. Geophys. Res. 88, 8379-8400.

Finkelstein, P.L. and Seilkop, S.K. (1981). Interpolation error and the spatial variability of acid precipitation, Seventh Conf. on Probability and Statistics in Atmospheric Sciences Am. Meteorol. Soc., Boston, Mass., pp. 206-212.

Madsen, B.C. (1982). An evaluation of sampling interval length on the chemical composition of wet-only deposition, Atmos. Envir. 16(10), 2515-2519.

Matheron, G. (1963). Principles of geostatistics, Econ. Geol. 58, 1246-1266.

Matheron, G. (1967). Kriging or polynomial interpolation procedures. Can. Inst. Mining Bull. 60, 1041-45.

MOI, 1982: Report of Work Group 3B on Emissions, Costs and Engineering Assessment, United States-Canada Memorandum of Intent on Transboundary Air Pollution.

Mueller, P.K. and Hidy, G.M. (1983). The sulfate regional experiment: report and findings, Vol. 3, Report EA-1901, ERT Inc. Concord, Mass.

Pack, D.H. (1980). Precipitation chemistry patterns: a two-network data set, Science 208, 1143-1145.

Parzen, E. (1964). An approach to empirical time series analysis, Radio Sci. J. Res., 68D, 551-565.

Reinsel, G., Tiao, G.C., Wang, M.N., Lewis, R. and Nychka, D. (1981). Statistical analysis of stratospheric ozone data for the detection of trends, Atm. Env. 15, 1569-1577.

Streets, D.G., Knudson, D.A. and Shannon, J. (1983). Selected strategies to reduce acidic deposition in the U.S. Env. Sc. Tech., 17, 474-485.

Samson, P.J., Moody, J.L., Kahl, J. and Keeler, J. (1983). Uncertainties in quantifying source-receptor relationships for atmospheric acids, In, Proc. NATO Adv. Study Inst. on the Chemistry of Multiphase Atmospheric Systems, Corfu, Greece (in press).

Thiessen, A.H. (1911). Precipitation averages for large areas, Mon. Wether Rev. 39, 1082-1084.

Tiao, G.C., Box, G.E.P. and Hamming, W.J. (1975). A statistical analysis of the Los Angeles CO data 1955-1972, J. Air Poll. Cont. Assoc. 25, 1129-1136.

Topel, L.E., Colovos, G., and Schwall, R. (1982), "Precisiion of precipitation chemistry measurements," pp. 197, Proceedings of APCA Specialty Conference on Atmospheric Deposition, Nov. 7-10, 1982, Detroit, Mich.

Wigley, T.M.L. and Jones, P.D. (1981). Detecting CO_2-induced climatic change, Nature 292, 205-208.

Young, J.W.S. and Shaw, R.W. (1983). A science-based strategy for the control of acidic deposition in North America. Proc. VI World Congress on Air Quality, Paris, May, 1983.

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Appendix 2

Detecting a Change in Wet Deposition due to a Change in the Strength of a Point Source

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The question to be addressed in this note is: how long is it necessary to make wet deposition measurements at a site in order to detect a change in the emission from a single point source within specified error bands? To answer this question, let us assume that the time series of wet deposition measurements behaves as shown in Figure 1. The deposition is a constant value D_p whenever the receptor "sees" the plume. If the "background" deposition at the sampler is assumed to be constant, D_p represents the excess over this value. The mean deposition at the sampler is:

$$D = fD_p \tag{1}$$

where f is the fraction of time the plume is detected by the sampler. To estimate f, assume that the plume has a constant width θ. Taking $\sigma_y = 0.1x$, θ becomes $0.1\sqrt{2\pi}$ radians or 15^o. Then f is approximately $\theta/2\pi$ or $\simeq 0.04$. The peak-to-mean ratio for our time series is then

$$D_p/D = 25.$$

Then it is easy to show that the ensemble variance is given by (Venkatram, 1979).

$$\sigma^2 = \langle (D(t) - \langle D \rangle)^2 \rangle = \langle D \rangle^2 \left[\frac{D_p}{\langle D \rangle} - 1 \right]. \tag{2}$$

In a recent paper, Sykes (1984) shows that the expected deviation between the time averaged deposition $\overline{D}^T$ and the ensemble average is given by

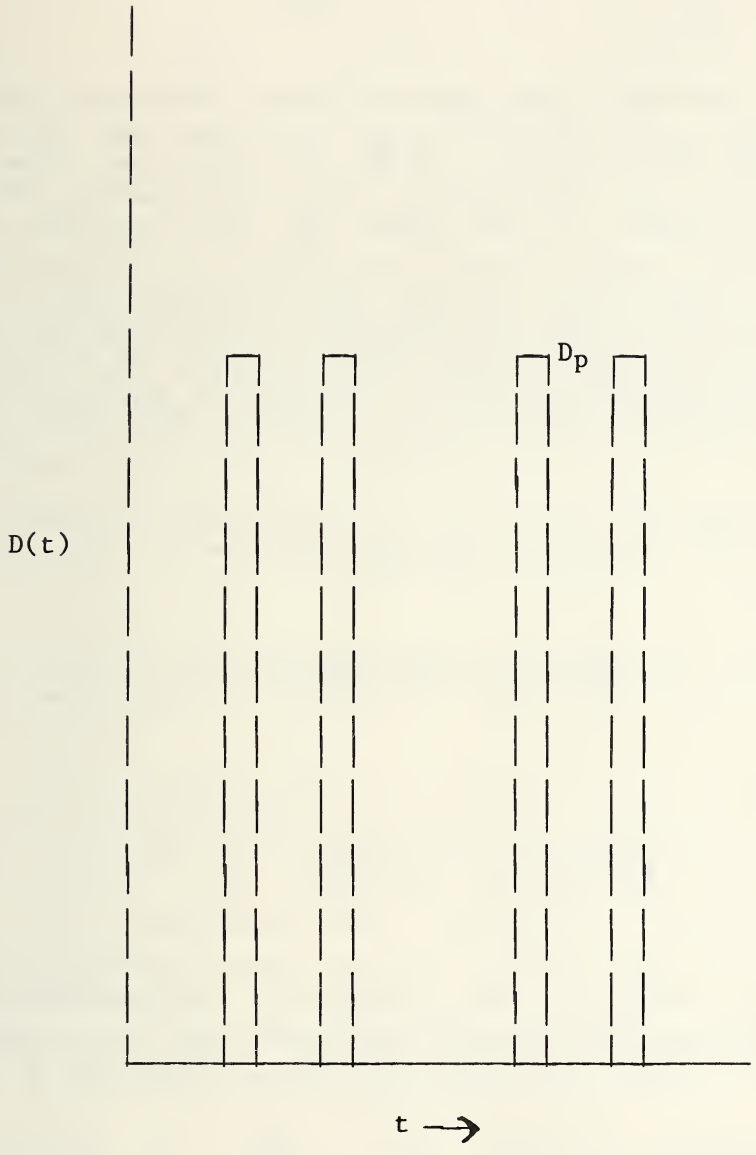

Figure 1: Idealized time series of wet deposition at a monitoring station.

$$\langle(\bar{D}^T - \langle D\rangle)^2\rangle = \frac{T_E}{T} \ln\left[1 + 2\frac{\sigma^2}{\langle D\rangle^2}\right] \langle D\rangle^2. \qquad (3)$$

In (3), T_E is the "average" time interval between deposition events at a given receptor. To estimate T_E, let us assume that the wind swings through 360° over 24 hours. As $\theta = 13°$, the plume spends 1 hour over a receptor. We know that it rains approximately 10% of the time. For convenience, we will assume that this translates into 4 hours of rainfall every 48 hours for a total of 730 hours over a year. Then over a period of 48 hours, the probability that it is raining when the plume is over the receptor is approximately 1/6. This means that the average interval between deposition events is (6 x 48) hrs = 12 days. We are now in a position to use Eq. (3) to estimate the time required to detect an emission change at the 95% confidence level.

If we assume that wet deposition is proportional to emission, detection at the 95% confidence level is equivalent to the statement

$$\langle D_n\rangle^2 \left[\frac{1}{r} - 1\right]^2 = 4\Sigma^2 \qquad (4)$$

where

$$\Sigma^2 \equiv \langle(\bar{D}^T - \langle D_n\rangle)^2\rangle, \qquad (5)$$

r is the ratio of the new emission rate to the old emission rate, and the subscript n refers to "new" deposition. Substituting (4) into (3), we arrive at the following expression for the averaging time T required to detect an emission change r at the 95% confidence level:

$$T = \frac{4T_E}{(1/r - 1)^2} \ln\left[\frac{2D_p}{\langle D_n\rangle} - 1\right]. \qquad (6)$$

Using our estimates for T_E and the peak-to-mean ratio, T (in days) is given by

$$T = 187/(1/r-1)^2. \qquad (7)$$

The following table give T for chosen values of r:

r	T (years)
0.9	4.2
0.5	0.5
0.2	0.03

Notice that T falls off very rapidly with r. It should be pointed out T is the time required to detect a change. It does not refer to the averaging time required to measure $\langle D_n \rangle$ within specified confidence limits. To illustrate this difference, let us assume that we want our measured value $\bar{D}^T$ to be within 10% of the ensemble average $\langle D_n \rangle$. Equivalently, we want

$$\Sigma / \langle D_n \rangle = 0.1.$$

Then from (3), we find that the averaging time required to achieve this measurement is $\sim$12 years. For a 20% error, the time required is 3 years.

These estimates of T are likely to be minimum values because we have not accounted for the "noise" introduced by other sources. Also, note that our estimate for T_E assumes an uniform distribution of rainfall. This cannot be justified for T less than a year. Therefore, caution has to be exercised in interpreting the preceding table.

References

Sykes, R.I., 1984. The variance in time-averaged samples from an intermittent plume. Atm. Envir. 18, 121.

Venkatram, A., 1979. The expected deviation of observed concentrations from predicted ensemble means. Atm. Envir. 13, 1547-1549.

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